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# RESEARCH MEMORANDUM

MEASURED EFFECTIVE THERMAL CONDUCTIVITY OF URANIUM  
OXIDE POWDER IN VARIOUS GASES AND GAS MIXTURES

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**NATIONAL ADVISORY COMMITTEE  
FOR AERONAUTICS**

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MEASURED EFFECTIVE THERMAL CONDUCTIVITY OF URANIUM OXIDE

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SUMMARY

As a part of an investigation of the effective thermal conductivities of powders, this study was undertaken to determine the effective conductivity of uranium oxide  $UO_2$  powder with a density of 59.5 percent of the density of the solid uranium oxide at temperatures between  $200^\circ$  and  $1500^\circ$  F. Helium, argon, and nitrogen gases and mixtures of helium-argon and xenon-krypton gases were introduced into the powder void, and the experimental effective thermal conductivities of the gas-powder mixtures were determined. All tests were performed at a gas pressure above the breakaway pressure, where the thermal conductivity is independent of pressure.

A method for determination of the relative thermal conductivity of the solid was devised from knowledge of the effective conductivity of the powder and the conductivity of the gases used in the two-phase system. This method was applied to the uranium oxide used in this investigation to determine its relative thermal conductivity through the  $200^\circ$  to  $1500^\circ$  F temperature range.

INTRODUCTION

Previous investigations of the thermal conductivities of magnesium oxide and stainless steel powders in various monatomic gases have resulted in a possible method of predicting the effective thermal conductivity of a gas-powder mixture from the properties of the solid and the gas (ref. 1). In reference 2, the effective thermal conductivity of uranium oxide with a void fraction of 0.37 was determined for helium and argon gases through the temperature range  $200^\circ$  to  $1250^\circ$  F. This investigation was proposed to obtain effective thermal conductivities for uranium oxide with a void fraction of 0.405 for xenon-krypton and helium-argon gas mixtures as well as helium, argon, and nitrogen gases. The temperature range was expanded to  $200^\circ$  to  $1500^\circ$  F.

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The test section was constructed by the NACA Lewis laboratory and filled with the uranium oxide powder by the General Electric Company.

## EXPERIMENTAL INVESTIGATION

### Apparatus

The apparatus used in this investigation is essentially the same as that described in reference 1 (fig. 1). Minor alterations, which are described in the following paragraphs, were incorporated into the design of the test section and the enclosing gas chamber.

Test section. - The test section consisted of two concentric tubes separated by the uranium oxide powder. The inner ceramic tube was heated electrically by a carbon rod, and the outer monel metal tube was cooled by convection.

The inner ceramic tube, with the wound end heaters and the platinum - platinum rhodium (10 percent) thermocouples stretched through the powder, was held in position by poured ceramic end plugs instead of machined disks as described in reference 1. The 1/4-inch-diameter filling tube and the end heater leads were brought out through these end plugs.

The thermocouple wires were coated with an insulating cement to electrically insulate the thermocouples from the uranium oxide powder. Because the added cement coating displaced a considerable portion of the usable powder volume, four groups of four, instead of five, radially positioned junctions were located in approximately equal increments across the central plane. The end heaters and the ceramic tube thermocouple leads were also cement-coated to eliminate the possibility of short-circuiting through the powder.

The main heating element, a 1/4-inch-diameter carbon rod, was inserted through the hollow ceramic tube case of the test section. Copper terminal blocks located at each end outside the test section insured proper electrical contact. Platinum end heaters were employed in conjunction with the carbon rod to prevent heat losses at the ends.

Gas chamber. - Thermocouple and electric power leads from the ends of test section were brought through the gas chamber wall by means of fittings that employed the crushing action of soapstone cones around the wire leads to effect a high pressure seal. The flange ends were sealed by a rubber O-ring recessed into a channel on the water-jacketed gas chamber.

### Procedure

The test section was filled through the filler tube with uranium oxide powder and vibrated intermittently with additions of powder for approximately  $1\frac{1}{2}$  hours. This procedure was repeated until the section was completely filled and packed to the maximum density. The density of the powder was calculated as 59.5 percent of the density of the solid uranium oxide (density of solid  $\text{UO}_2$  assumed to be 10.7 g/cc), giving a void volume in the test section of 40.5 percent. The particle sizes of the powder are given in table I.

The positions of the thermocouple junctions distributed through the powder were measured from X-ray photographs of the test section after it was filled. Data were obtained for helium, argon, and nitrogen gases and mixtures of helium-argon and xenon-krypton gases over a range of average powder temperatures and pressures given in the following table:

Gas <sup>a</sup>	Pressure, lb/sq in. abs	Temperature, °F
Helium	59.3 to 136.9	226 to 1124
Argon	44.3 to 94.4	195 to 1450
Nitrogen	49.3 to 83.3	339 to 1161
Xe/Kr, 4.898	18.78 to 74.3	235 to 1400
He/A, 1.857	47.3 to 84.3	225 to 1014
He/A, 0.953	39.4 to 96.3	169 to 875
He/A, 0.333	46.3 to 84.3	215 to 1062

<sup>a</sup>Gas mixtures given as volumetric ratio.

The method of obtaining the data is presented in reference 1. Only the portion of the power input to the carbon rod between the end heaters was used in thermal conductivity calculations of the powder. Continually higher temperature runs were made after cross-checking the lower temperatures until the thermocouples failed.

### Reduction of Experimental Data

The derivation of equations used in calculating values of thermal conductivity of the powder from the experimental data may be found in reference 1, together with a correction method for the eccentricity of the tube. Thermal conductivities of the gases and gas mixtures used in this investigation are given in figure 2 using data from references 3 to 5. Calculations of the conductivity of the gas mixtures were based on the Enskog equation (ref. 4) for the thermal conductivity of a binary mixture of gases. For the xenon-krypton mixture, this equation reduces to

$$k_{\text{Xe/Kr}}(4.898) = 0.0978 k_{\text{Kr}} + 0.842 k_{\text{Xe}} + 0.1265 (k_{\text{Kr}} k_{\text{Xe}})^{1/2} \quad (1)$$

Three mixtures of helium and argon gases were studied with helium-argon ratios of 1.857, 0.953, and 0.333. The conductivities of these gas mixtures were calculated in the same manner as the xenon-krypton mixture and were plotted in figure 2(a). Equations for the thermal conductivity of these mixtures are

$$k_{\text{He/A}}(1.857) = 0.563 k_{\text{He}} + 0.3315 k_{\text{A}} + 0.0583 (k_{\text{He}} k_{\text{A}})^{1/2} \quad (2)$$

$$k_{\text{He/A}}(0.953) = 0.398 k_{\text{He}} + 0.491 k_{\text{A}} + 0.0609 (k_{\text{He}} k_{\text{A}})^{1/2} \quad (3)$$

$$k_{\text{He/A}}(0.333) = 0.186 k_{\text{He}} + 0.736 k_{\text{A}} + 0.0428 (k_{\text{He}} k_{\text{A}})^{1/2} \quad (4)$$

## RESULTS AND DISCUSSION

### Experimental Effective Conductivities

The experimental values of the effective thermal conductivity of uranium oxide powder between 200° and 1500° F in an atmosphere of various gases and gas mixtures are presented in figure 3. The void fraction occupied by the gas was 0.405 in all runs. Inasmuch as the effective thermal conductivity of the gas-powder mixture varies somewhat at lower gas pressures, depending on the temperature, the characteristics of the gas, and the relative dimensions of the gas space (as was shown to be true in ref. 1), all data presented were obtained at pressure levels high enough for conductivity to be independent of pressure. Determination of the magnitude of these pressure levels is subsequently discussed.

Thermal conductivities of uranium oxide powder in argon, nitrogen and helium are plotted in figure 3. The thermal conductivity of the powder in argon and helium increases to a maximum with temperature in much the same manner as that of the uranium oxide powder with a void fraction of 0.37 presented in reference 2. The effective conductivity of the powder in nitrogen exhibits a slight decrease to a minimum value with an increase in temperature. Only six data points were determined using this gas, in view of the possibility of reduction of the powder at elevated temperatures.

In figure 3(b), the variation of the effective thermal conductivity of uranium oxide powder with a xenon-krypton gas mixture (ratio 4.898:1)

in a temperature range from 200° to 1500° F has been plotted from the data. A reference plot, that of figure 2(b), has been inserted to facilitate comparison of the conductivities of the xenon-krypton gas mixture to the conductivities of the two gases.

Three mixtures of helium and argon gases were studied with helium-argon ratios of 1.857, 0.953, and 0.333 (fig. 3(a)). The effective conductivities of the powder with the helium-argon gas mixtures follow the characteristic effective conductivities for the powder with the component gases. Above 500° F, the effective thermal conductivity of the powder with helium and argon gases and gas mixtures appears to be independent of temperature. Effective thermal conductivity for temperatures greater than 500° F is plotted against argon-helium mixture composition in figure 4. A linear relation appears to exist between the experimental effective thermal conductivities and helium-argon gas mixture composition.

In this investigation, the temperatures of the uranium oxide powder reached values 200° F higher than the temperatures obtained for the same powder with a void fraction of 0.37 investigated in reference 2.

#### Effect of Pressure on Conductivity

The effect of gas pressure on the effective thermal conductivity of a gas-powder mixture was shown in reference 1, by using the kinetic theory of gases, to be a function of the Knudsen number (the ratio of the mean free path of the gas molecules to a characteristic dimension of the gas spaces). Because the size of the gas spaces is of the same order of magnitude as the size of the particles, the characteristic dimension is taken as the weighted mean sieve size of the particles, and is calculated from the data in table I, resulting in 0.0002717 foot for the uranium oxide powder. The predicted pressure (breakaway pressure) below which the effective thermal conductivity begins to vary appreciably is given by the following equation from reference 1:

$$p_b = 1770 \times 10^{-24} \frac{t}{s^2 l_s} \quad (5)$$

where

$p_b$  breakaway pressure, lb/sq ft abs

$t$  gas temperature, °R

$s$  molecular diameter determined from viscosity, ft

$l_s$  weighted mean sieve size required to retain particles, ft

The constant in this equation was obtained from experimental data in reference 1. The molecular diameter  $s$  in feet is  $7.22 \times 10^{-10}$  for helium (ref. 6),  $9.45 \times 10^{-10}$  for argon,  $10.33 \times 10^{-10}$  for nitrogen,  $12.11 \times 10^{-10}$  for krypton, and  $13.19 \times 10^{-10}$  for xenon (ref. 7). The molecular diameters for the mixtures were calculated, based on the mole fraction  $n$  through the addition of  $n_1 s_1$  and  $n_2 s_2$ , as shown in the following table:

1	2	$n_1$	$n_2$	$n_1 s_1$	$n_2 s_2$	$s_{\text{mixture}}$
Xe	Kr	0.8305	0.1695	$10.954 \times 10^{-10}$	$2.053 \times 10^{-10}$	$13.007 \times 10^{-10}$
He	A	.650	.350	4.693	3.308	8.001
He	A	.4885	.5115	3.527	4.834	8.361
He	A	.250	.750	1.805	7.088	8.893

From equation (5) and the constants given in the preceding table, the breakaway pressure was calculated for each gas and gas mixture through the temperature range  $200^\circ$  to  $1500^\circ$  F. The pressure of the gas in the test section was maintained above this calculated value for all runs in this investigation.

#### Calculation of Relative Thermal Conductivity of Solid Uranium Oxide

The effective thermal conductivity of a gas-powder mixture has been shown in reference 1 to depend principally on the thermal conductivity of the gas, the thermal conductivity of the solid, and the void fraction of the powder. Therefore, if a powder is packed to a fixed density or void fraction, the resulting effective thermal conductivity  $k_{\text{eff}}$  is a function of the thermal conductivities of the gas  $k_g$  and the solid  $k_{\text{UO}_2}$ , or, for this investigation,

$$\frac{k_{\text{eff}}}{k_g} = f \left( \frac{k_{\text{UO}_2}}{k_g} \right) \quad (6)$$

Figure 5(a) is a plot of  $k_{\text{eff}}/k_g$  against the conductivity of the gas for the uranium oxide powder used in this investigation through a temperature range of  $200^\circ$  to  $1500^\circ$  F. Values for this plot were obtained from the faired curves of figures 2 and 3.

From equation (6), for a constant value of  $k_{eff}/k_g$ ,  $k_{UO_2}$  is proportional to  $k_g$ . To obtain the relative effect of temperature on the thermal conductivity of the solid from this proportionality, an arbitrary reference temperature must be assumed. Considering 600° F a reference temperature, it follows that, for equal values of  $k_{eff}/k_g$ ,

$$\frac{k_g}{k_{g,600}} = \frac{k_{UO_2}}{k_{UO_2,600}} \quad (7)$$

From figure 5(a), values of  $k_g$  at temperatures from 200° to 1500° F were obtained for constant values of  $k_{eff}/k_g$ . Using a reference temperature of 600° F, values of  $k_g$  from figure 5(a), and equation (7), the relation of  $k_{UO_2}/k_{UO_2,600}$  to temperature  $T$  was plotted (fig. 5(b)) for four values of  $k_{eff}/k_g$ . One curve can be drawn which represents all fixed values of  $k_{eff}/k_g$ . This was to be expected, since the conductivity of the solid should be a function only of the temperature. The negative slope of this curve suggests that the thermal conductivity of solid uranium oxide decreases with increasing temperature. This variation is in agreement with previous observations in reference 2.

Since figures 2, 3, and 5(b) give the conductivity of the gases, the effect conductivity of the powder, and the relative conductivity of the solid, a plot of  $k_{eff}/k_g$  against  $k_{UO_2}/k_g$  as suggested by their relation in equation (6) might be possible if the thermal conductivity of the solid uranium oxide of this experiment at some reference temperature was known. In the absence of this information,  $k_{eff}/k_g$  (from experimental data, fig. 3, and  $k_g$  from fig. 2) was plotted against  $k_{UO_2}/k_g(k_{UO_2,600})$  for corresponding temperatures in figure 6(a). The abscissa may be altered to any desired reference temperature by dividing by the ratio  $k_{UO_2,T_r}/k_{UO_2,600}$  from figure 5(b) for that temperature  $T_r$ .

It was noted in reference 2 that for two test conditions using an identical material and gas at identical temperatures but different void fractions, the effective thermal conductivities of the gas-power mixtures tend to have lower values for greater void fraction. From the data of reference 2 on the effective conductivity of uranium oxide (void fraction 0.37) figure 6(b), which corresponds to figure 6(a), was developed from the knowledge of the relative thermal conductivity of solid uranium oxide. Though the data for both of these plots deviate slightly from any single curve relation, it may be seen that the effective conductivity for the uranium oxide powder tends to have lower values for the greater void fraction.



The procedure used here for determination of the relative conductivity of a solid material is applicable for the determination of the thermal conductivity of any solid material if its conductivity at any one temperature has been established and knowledge of the effective conductivity of the powder in several gases or gas mixtures is known for the desired temperature range. Though the procedure followed here will apply to any system, it should be remembered that figure 5 is unique to this investigation.

#### SUMMARY OF RESULTS

From the investigation of the effective thermal conductivity of uranium oxide powder in helium, argon, and nitrogen gases, and mixtures of helium and argon, and xenon and krypton, the following results were obtained:

1. For temperatures above 500° F, the effective thermal conductivity of the uranium oxide powder in various gases was substantially independent of temperature.
2. An increase in the thermal conductivity of the gas or the gas mixture increased the effective conductivity of the gas-powder mixture.
3. All the data on effective conductivity of the uranium oxide powder in the various gases could be correlated into a single curve by means of the relation that the ratio of the effective conductivity to the gas conductivity is a function of the ratio of the conductivity of solid uranium oxide to the gas conductivity for a given percentage of void space in the compact.

Lewis Flight Propulsion Laboratory  
National Advisory Committee for Aeronautics  
Cleveland, Ohio, December 10, 1954

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TABLE I. - PARTICLE SIZE OF POWDER

Mesh Tyler sieve number	Sieve opening, <sup>a</sup> <sub>12</sub> S, in.	Average amount of powder retained on sieve <sup>a</sup> <sub>100</sub> f, percent	Line number, <sup>a</sup> n
100	0.0059	0	1
150	.0041	59.0	2
Through 325	Assumed .000748	41.0	3

<sup>a</sup>See ref. 1.

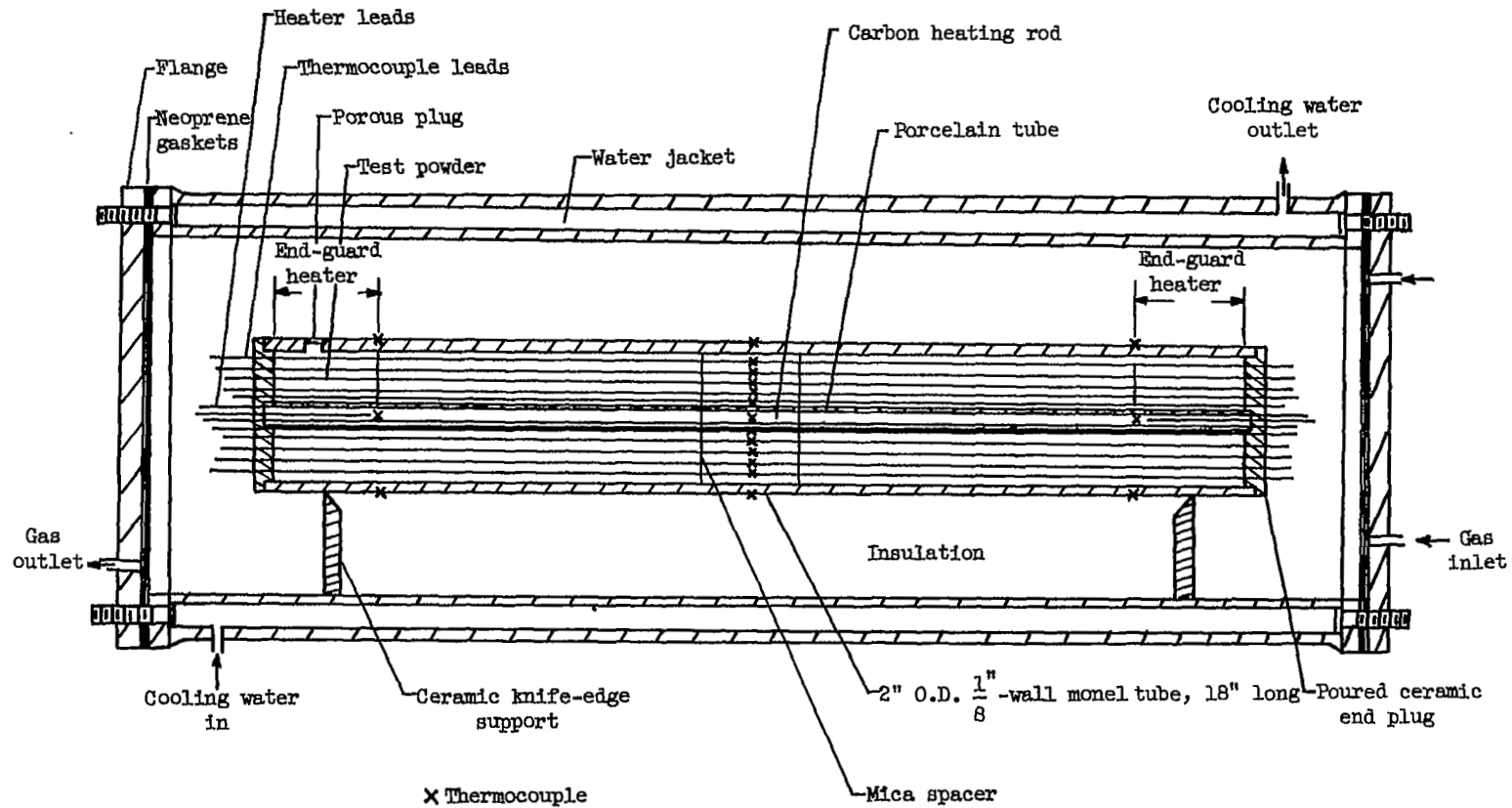
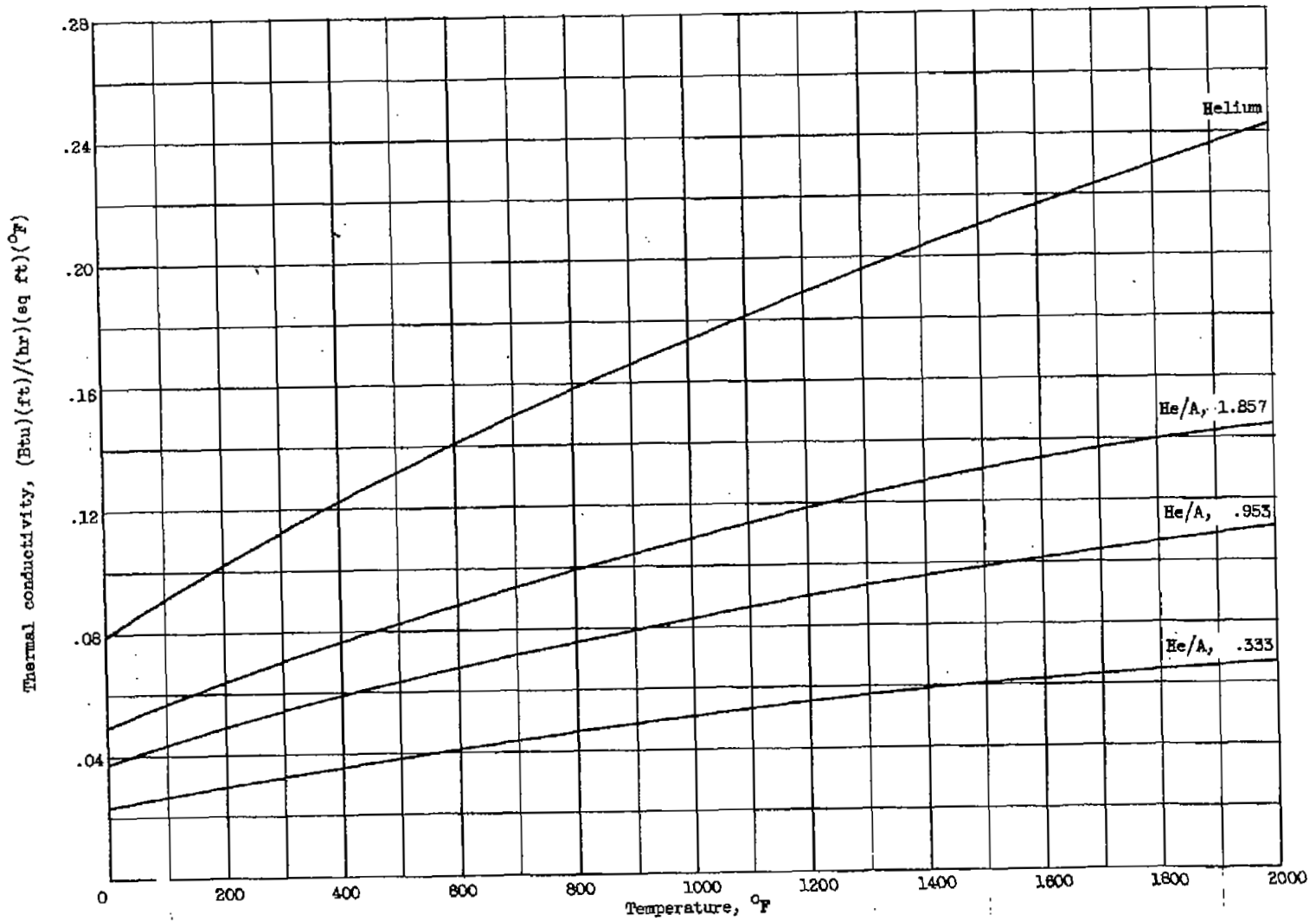
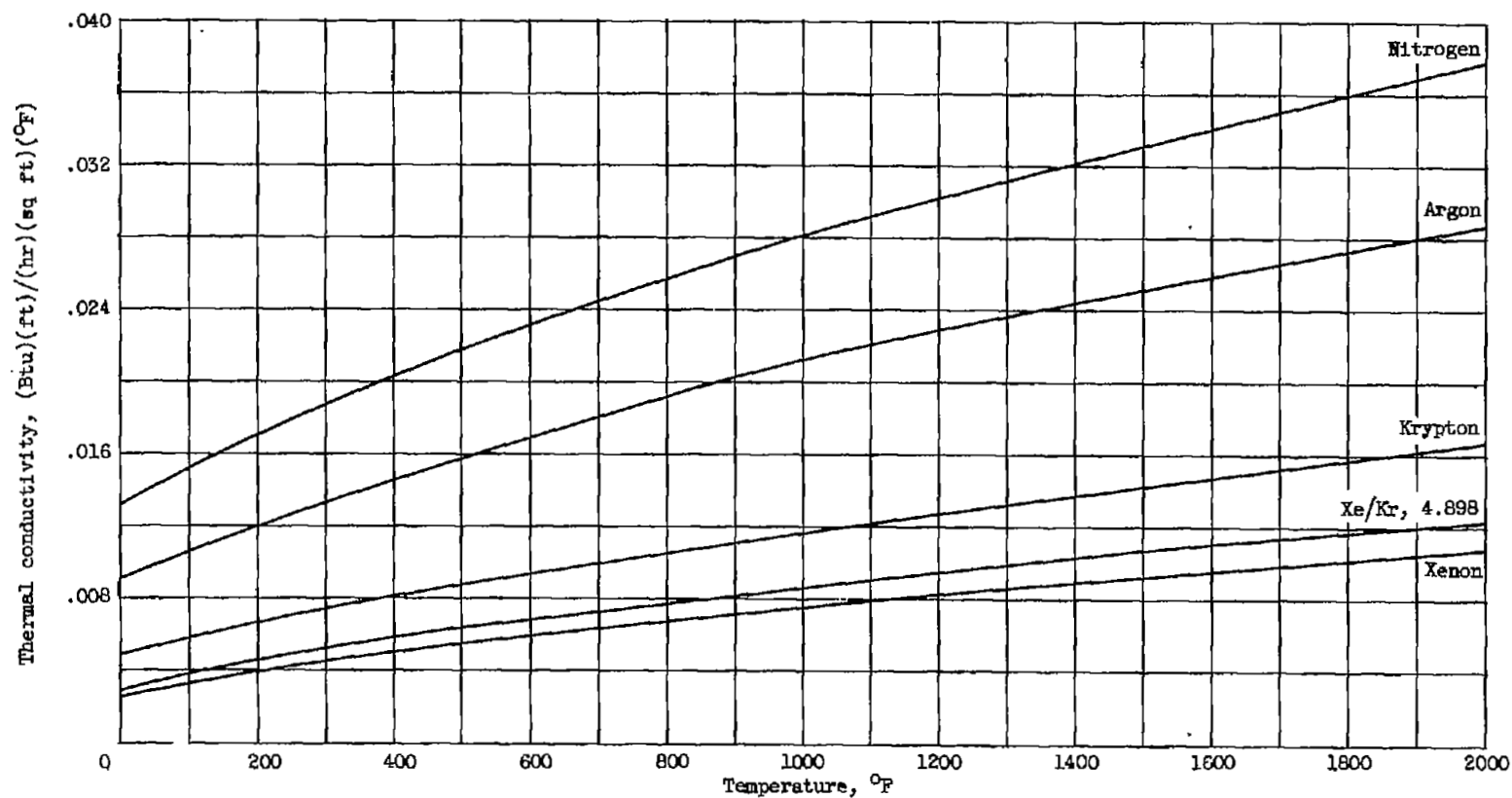


Figure 1. - Schematic drawing of test section and gas chamber used in determination of thermal conductivities of powders in various gases and gas mixtures.



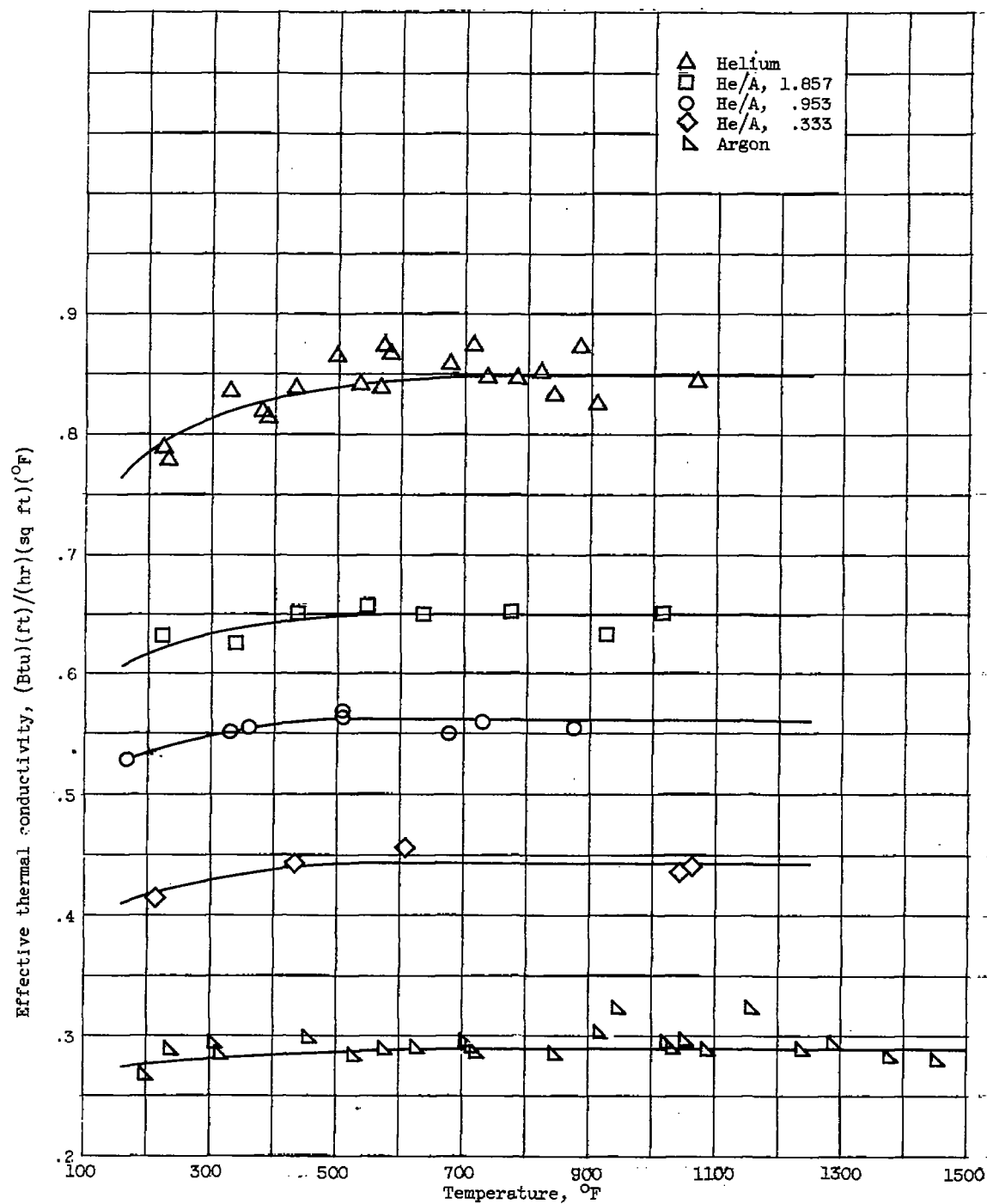
(a) Helium from reference 2, helium-argon mixtures from equations (2) to (4) and reference 4.

Figure 2. - Reference plot for the thermal conductivities of gases.



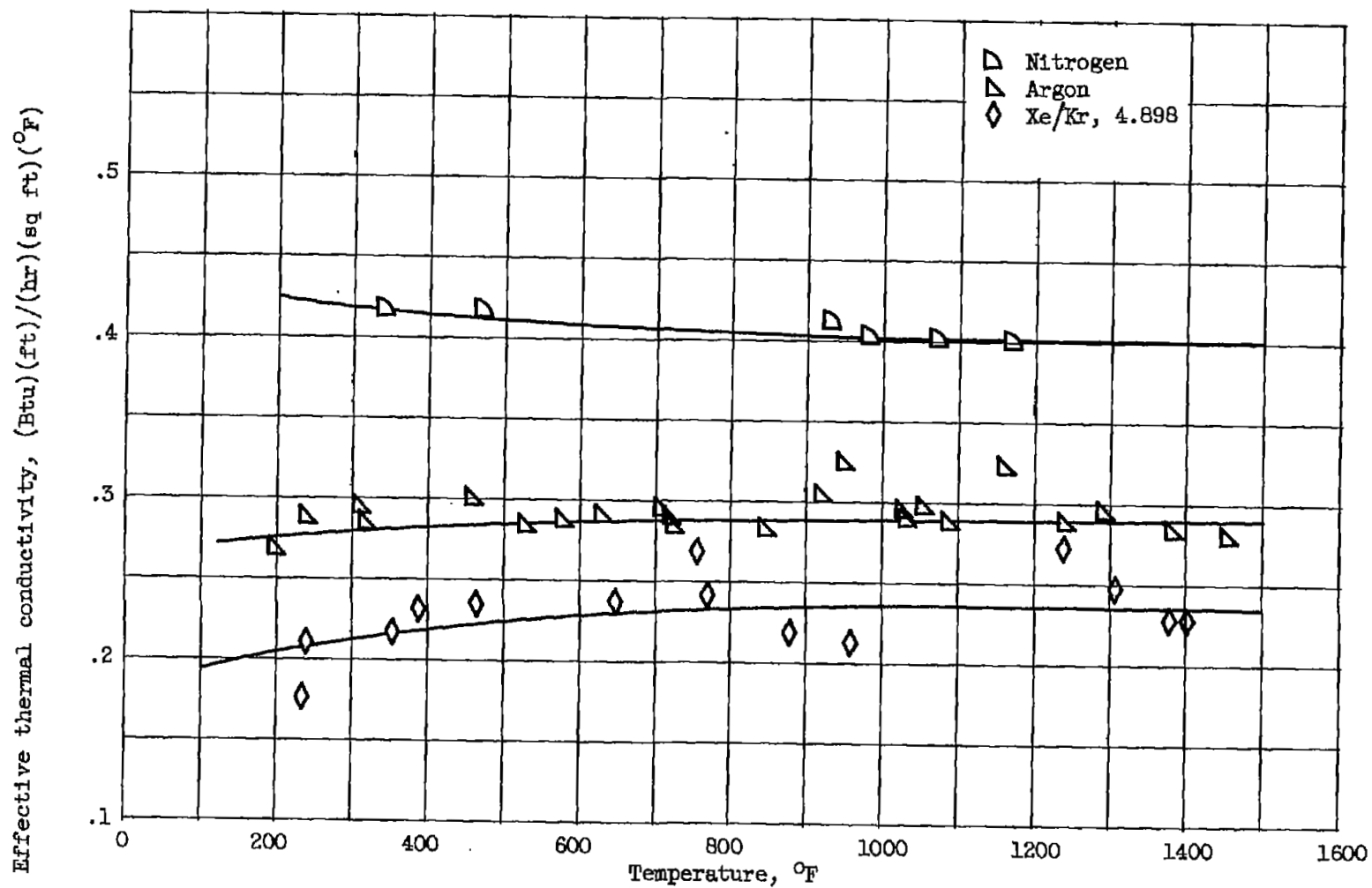
(b) Argon from reference 3, nitrogen from reference 4, xenon and krypton from reference 5, and xenon-krypton mixture from equation (1) and reference 4.

Figure 2. - Concluded. Reference plot for the thermal conductivities of gases.



(a) Helium, argon, and helium-argon mixtures.

Figure 3. - Experimental thermal conductivities of uranium oxide powder in various gases.



(b) Nitrogen, argon, and xenon-krypton mixtures.

Figure 3. - Concluded. Experimental thermal conductivities of uranium oxide powder in various gases.



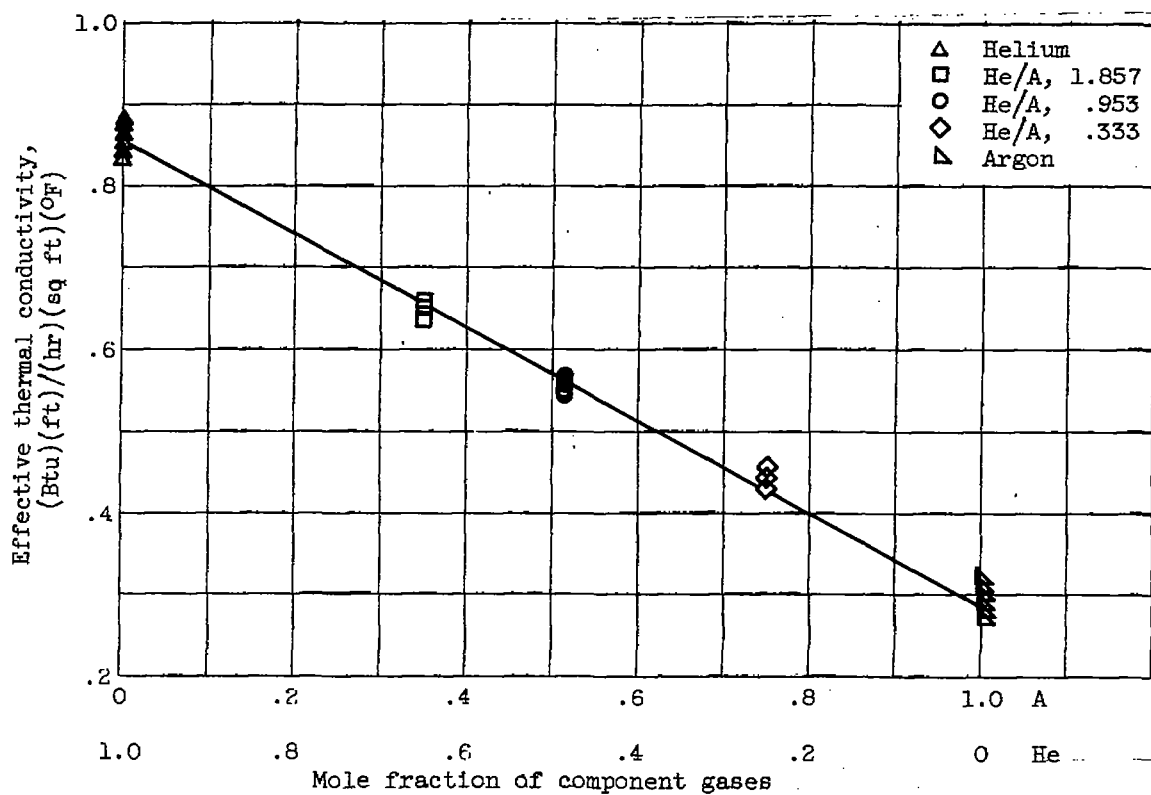
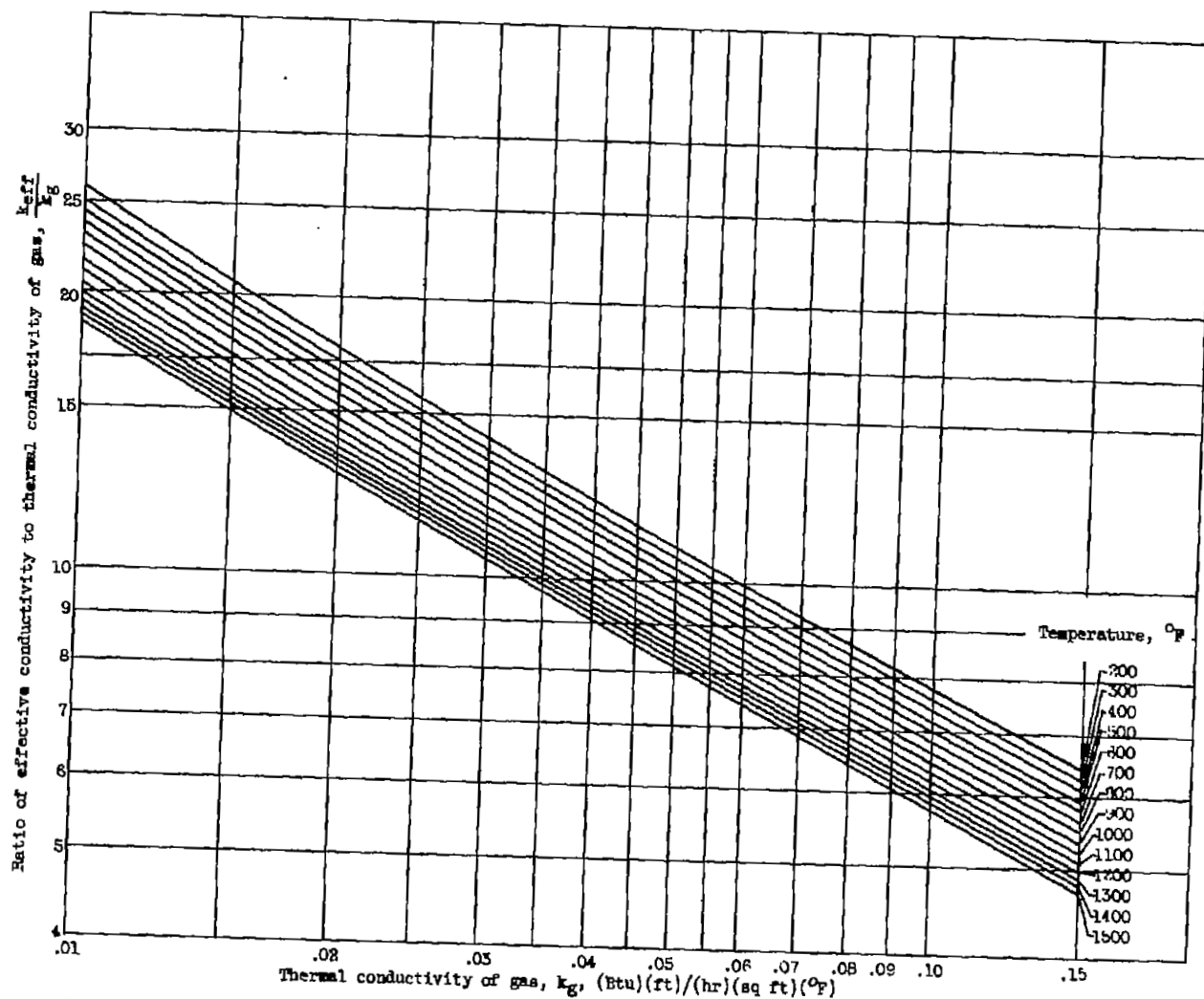
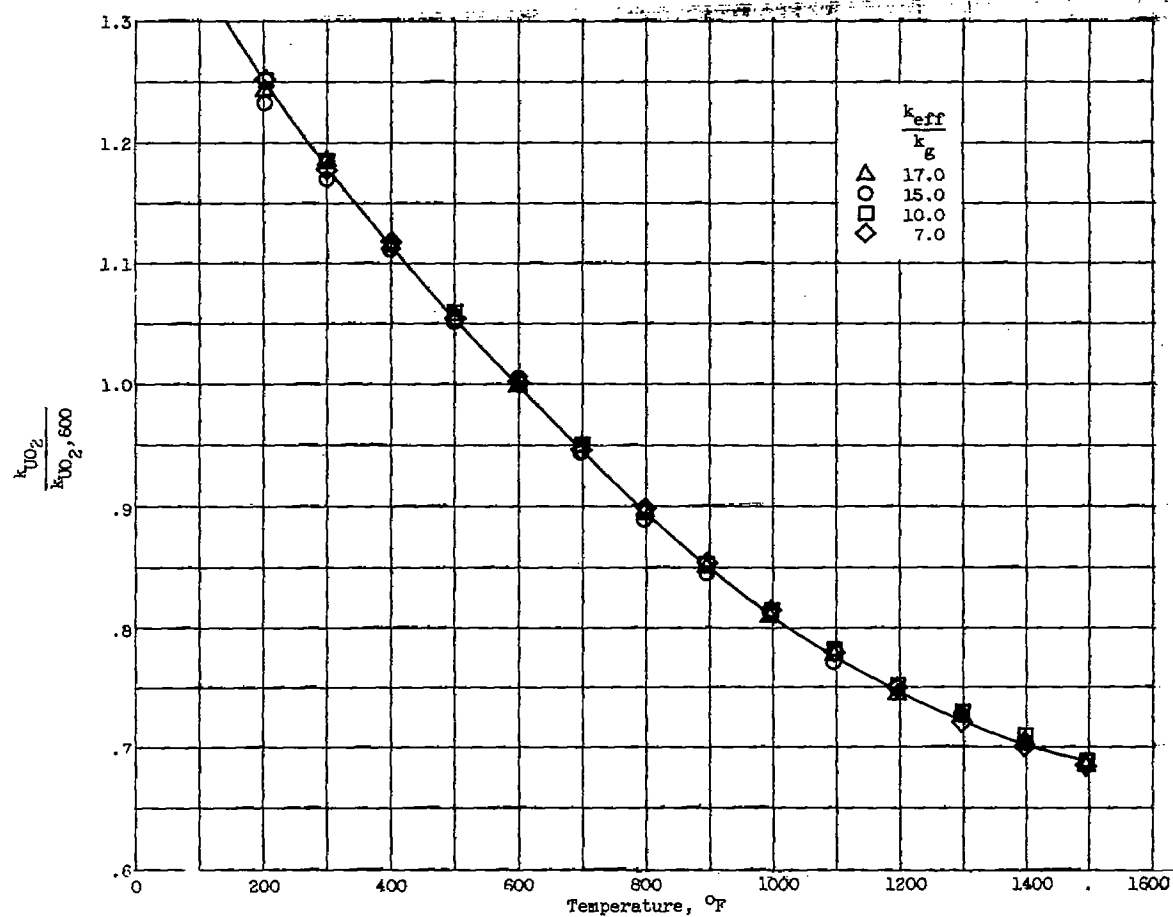


Figure 4. - Effect of gas mixture composition on experimental effective thermal conductivity using data at temperatures above 500° F where the effective conductivities appear independent of temperature.



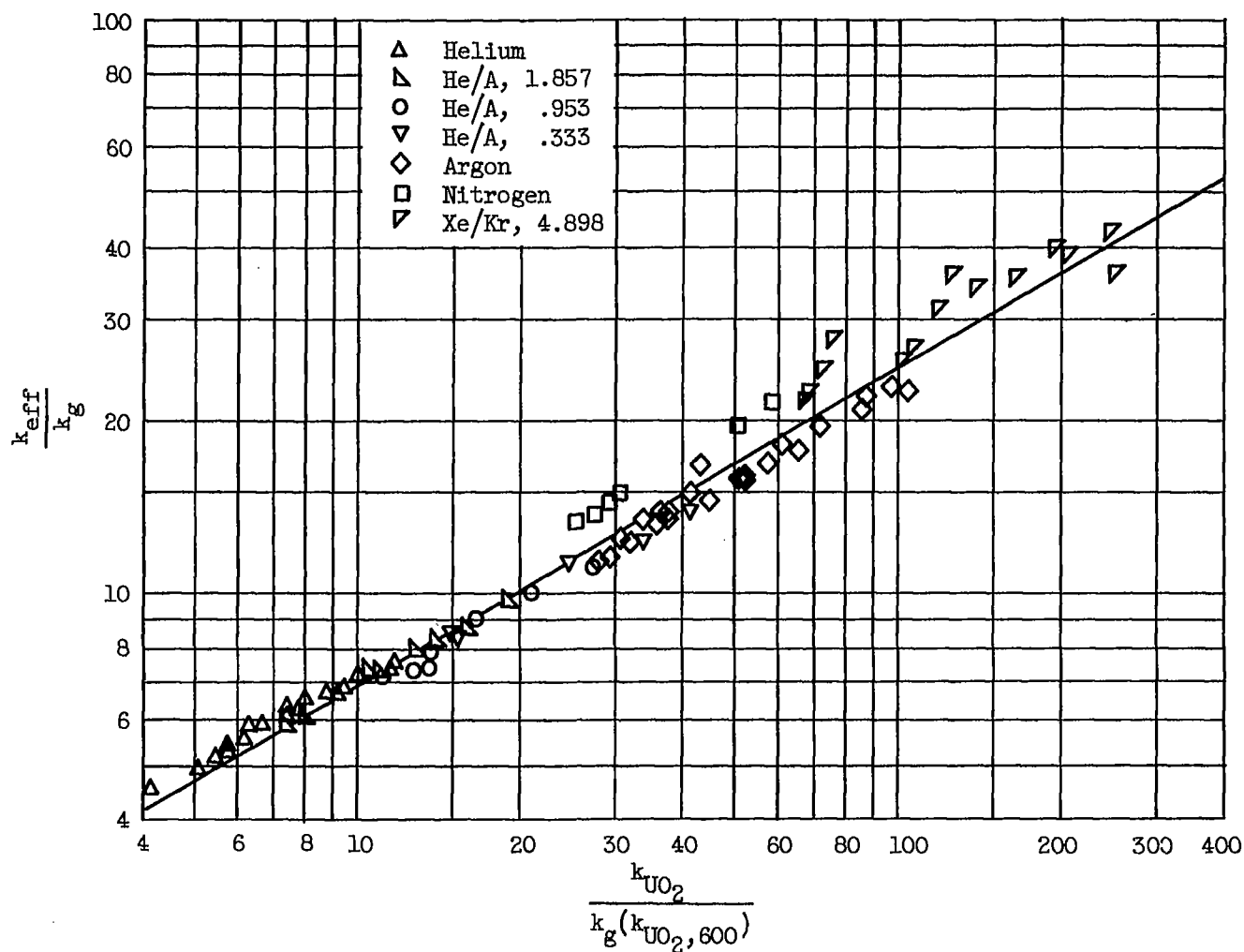
(a) Ratio of effective conductivity of the powder to thermal conductivity of the gas against thermal conductivity of the gas at various temperatures.

Figure 5. - Calculation of relative thermal conductivity of solid uranium oxide.



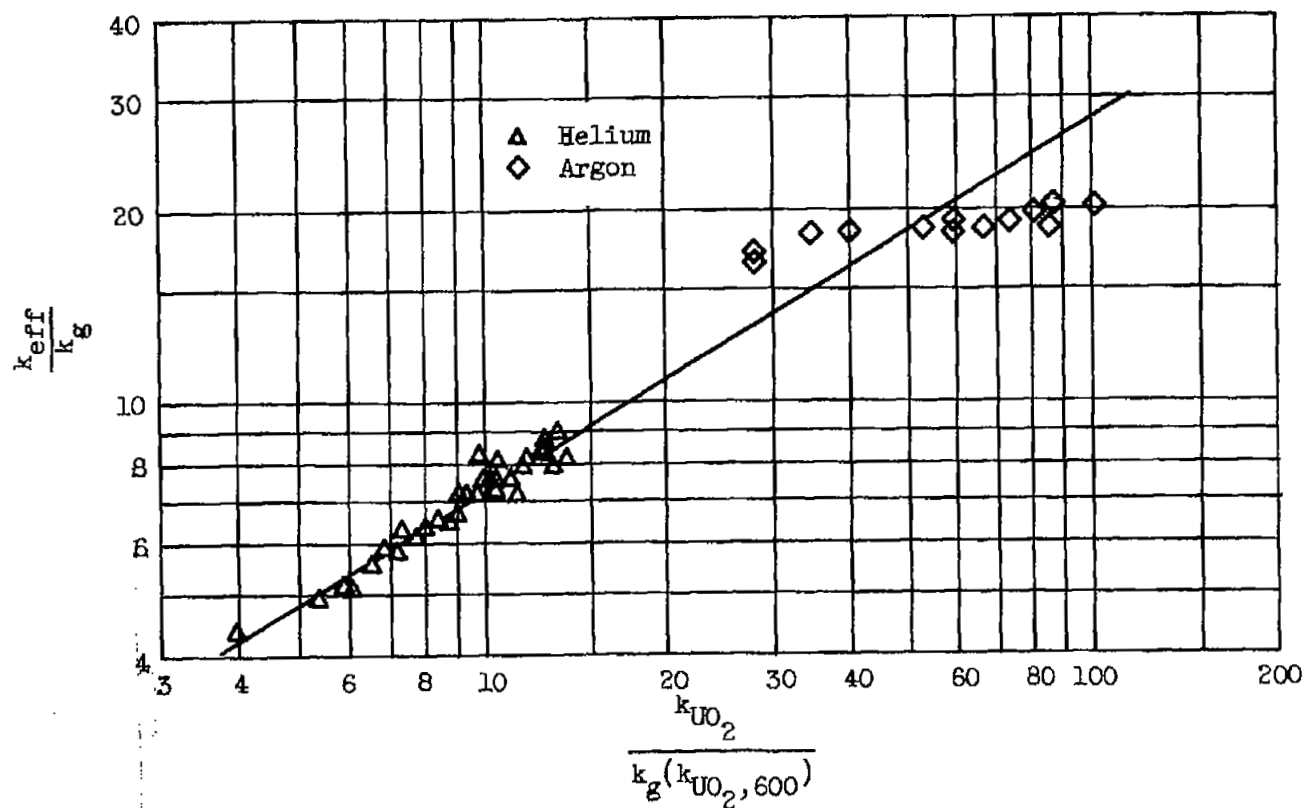
(b) Relative thermal conductivity of solid uranium oxide from 200° to 1500° F.

Figure 5. - Concluded. Calculation of relative thermal conductivity of solid uranium oxide.



(a) From experiment data on various gases and gas mixtures for void fraction of 0.405.

Figure 6. - Relation of conductivity of the gas, experimental effective conductivity, and relative thermal conductivity of the solid for uranium oxide.



(b) From experimental data for a void fraction of 0.37 (ref. 2).

Figure 6. - Concluded. Relation of conductivity of the gas, experimental effective conductivity, and relative thermal conductivity of the solid for uranium oxide.